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Butanol production draw from lignocellulosic residues under the biorefinery approach: Bibliometricalanalysis*

Producción de butanol a partir deresiduos lignocelulósicos bajo el concepto de biorrefinería: Análisis bibliométrico

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ABSTRACT

*With a world market that is around 4,2 (10^{12}) US$/year (3 Mton/year) and a growing projection of use, butanol is a compound that has more advantages as biofuel, than ethanol. Using the strategy of bibliometric analysis, this work looks to generate an integral vision of the determining research aspects concerning butanol through the combination of different keywords related to it. The databases used for the study, where those available in Scopus and Web of Science platforms, related to butanol in the VOSViewer during the period 1984-2020 and the first semester of 2021. It was also found that butanol production from lignocellulosic material does not yet show good yields and process intensification for an economically and environmentally acceptable biorefinery concept. Deficiencies in fermentation are a *Proyecto investigación origen: Becas Crédito-Condonable Formación Doctoral "Bicentenario"-Corte 1 (2020-2024). Financiación Beca Doctoral, Ministerio de Ciencia, Tecnología e Inovación- Convenio Colfuturo (2020). Colombia. Ejecución: Universidad del Valle; Grupo de investigación en Biocombustibles y Biorrefinerías GRUBIOC.*

KEYWORDS:
Biofuel; Biomass; Fermentation; Date bases; Biotechnology; Integral vision; Yields; Concentrations; Opportunities; Environmental; Energy.

RESUMEN

El butanol, es un compuesto que tiene más ventajas como biocombustible que el etanol, con un mercado mundial que está alrededor de 4,2 (10^{12}) US$/año (3 Mton/año), y una proyección de uso cada vez más creciente. Por esta razón, en el presente trabajo, bajo la estrategia de un análisis bibliométrico, se combinaron diferentes palabras claves relacionadas con el butanol, para generar una visión integral de los aspectos investigativos determinantes. Las bases de datos utilizadas, fueron las disponibles en las plataformas de Scopus y Web of Science, relacionada con el butanol en el VOSViewer durante el periodo 1984 a 2020 y el primer semestre de 2021. Se encontró que la producción de butanol a partir de material lignocelulósico, no muestra aún buenos rendimientos e intensificación de procesos, para un concepto de biorrefinería económica y ambientalmente aceptable. Un cuello de botella, son las deficiencias en la fermentación, por la falta de microorganismos más productores y tolerantes a altas concentraciones de butanol.

PALABRAS CLAVE:
Biocombustible; Biomasa; Fermentación; Bases de datos; Biotecnología; Visión integral; Rendimientos; Concentraciones; Oportunidades; Ambiental; Energia.

INTRODUCTION

Fossil fuels have increased pollutant emissions over time. The depletion of reserves has made their production neither sustainable nor viable. Biofuels, as a renewable energy source, have gained global interest due to their advantages as a renewable biomass, which are still being consolidated and require further research. The commitment of all sectors to promote biofuels is essential to meet the world's energy demand (Saini *et al*., 2019; Martínez *et al*., 2020).

Biofuels from lignocellulosic biomass represent an opportunity for the energy sector. Theproduction source is waste from agro-industrial processes, which can be valorized to have other products, which help to take care of the environment. These opportunities are possible as research offers solutions to logistical and technological problems (Wang *et al*., 2021; Hao *et al*., 2021).

The academic and scientific communities raise the need to have detailed studies of the factors that significantly influence the feasibility of the processes. Establish the evolution ofmethodologies and technologies. Elements of interest to identify through exhaustive studies are scaling, exergy analysis, process integration, economic, social and environmental indicators. There are experimental data that validate the obtaining of several products of commercialinterest, for example, butanol, which could strengthen the biofuels market (Martinez *et al*., 2020; Devasia *et al*., 2021; Brandt *et al*., 2021).

The production of butanol by traditional chemical and biotechnological methods is expensive (Trindade y Santos, 2017; Lee *et al*., 2008; Cheng *et a*l., 2019). The estimated value in the worldmarket is around 2,8 million tons/year, with 3,2 % growth/year distributed in 4,2 (10^{12}) US$/year (Durán Padilla, 2015). Future possibilities focus on improving the process, implementing the use of more available and abundant raw materials (Rajesh Kumar & Saravanan, 2016; Shibata *et al*.,2020; Callegari *et al*., 2020).

Others consist of improving energy integration unit operations (Silva-Lora *et al*., 2015), efficient use of water, intelligent control systems (Yasnitsky & Gladkiy, 2020). In this sense, a more efficient, profitable and environmentally sustainable process would be provided, by maintaining the production of main, secondary and by-products with added value in closed cycles *(*Shibat*a* et*a*l., 2020).

Sugarcane harvest residues, as a material, have the possibility of being used to obtain butanol, involving a biorefinery scheme (Mariano *et al*., 2013; Michailos *et al*., 2016; Mandegari, *et al*., 2018; Cimino *et al*., 2018; Magalhães *et al*., 2018; Cheng *et al*., 2019). The lignocellulosic source product would be more competitive than the one obtained by the normal chemical route. Gasoline engines accept up to 30 % ethanol, but butanol has better properties (under normal conditions boiling point of 118 ºC, melting point of -89 ºC, density of 0,816 g/m^3, lower calorific value of 34 MJ/kg, energy density of 29,2 MJ/L and heat of vaporization of 0,43 MJ/kg) (Trindade & Santos, 2017; Da Silva Trindade and Dos Santos, 2018). Studies of butanol (90 %) and gasoline (10%) blends have shown that the combustion process can be effectively controlled, making it very easy to use (Lu *et al*., 2021).

The technical feasibility of butanol production from biomass sources, with a biorefinery approach, can be analyzed with a global vision considering the data provided by the bibliometric tool (Pritchard, 1969) (Moral-Muñoz *et al*., 2020). The fundamental concepts and elements of bibliometrics are based on data from institutions and countries related to academic productivity, research contributions and funding.

Bibliometric techniques are of interest as a tool that contextualizes a specific field of research. This exhaustive study articulates activity and relationship indicators that avoid subjective interpretations. Through mathematical and engineering methodologies, they generate reliable statistics. These are inputs for the initiation of new research advances. And useful in the analysisof academic and scientific reports (Moral-Muñoz *et al*., 2020).

The elaboration of this type of documents is taken from the information published in journals and stored in platforms. The databases are then analyzed by exchanging and mixing keywords, which can be mapped graphically, such as VOSviewer (Moral-Muñoz *et al*., 2020). The purpose of this work was to evaluate butanol production with biomass sources and with a biorefinery approach, using the bibliometric strategy.

METHOD

The methodology for the bibliometric analysis was built mainly taking as a reference, the tools and methodologies researched and published by Moral-Muñoz, J.A. *et al*. (2019). And it was completed with the sources referenced in this study. The common methods defined are the systematic review of the topic of interest through database. Then definition of the limits and search equations. Then continue with the bibliometric analysis and finally the processing of the information using software.

The methods use activity indicators to generate files that are then analyzed with other software. The indicators allow the identification of themes in a particular field, co-words, co-occurrence and minimum group size. As well as the statistics of the evolution of the field of study and by means of software to generate the co-citation networks, authors and journals with the highest number of references.

Scopus and Web of Science databases were used as bibliographic sources for the different analyses. A first exploration consisted of a general review with the keyword "butanol" for the first half of 2020. Based on that, the review was further refined using search equations with more terms and connectors: (sugarcane OR biorefinery OR aspen AND butanol) AND (EXCLUDE (PUBYEAR, 2020), also using them independently for each platform. Then, a new search was executed, using the same strategy and, including documents from 2020. Considering the constant adjustment that the platform undergoes, as the year passes to reevaluate the initial information.

The evolutionary period of evaluation was from 1984 to 2020, involving set and subsets of wordsrelated to lignocellulosic biomass and biorefineries, under the global butanol landscape. Special consideration was given to the information considered to have the highest citation, relevanceand recency. Initially, 1.080 documents were obtained with Scopus in all areas, which were progressively reduced to 458, 396, 187 and 150 according to the combination and connectors used. Similarly, with Web of Science (WoS) platform, 8.776 records were initially generated, leaving 458 in each of the platforms. In CSV Excel (Scopus) and Plaint Text (WoS) formats, the information was exported and saved in separate folders to facilitate the mapping of relationships between keywords in VOSviewer software, using the "Reference manager files" command.

With both databases, the documents were classified by age and citation. On the other hand, the interest of the topic; the progress of the technology and its trends were evaluated. Using the "Analyze search result" option, statistics of interest were obtained, referring to documents by year, area of knowledge, type of documents, countries, territories, author, affiliation, funding sources and journals. Finally, with the results obtained, the productivity, state of the art and evolution of lignocellulosic butanol research were analyzed.

RESULTS

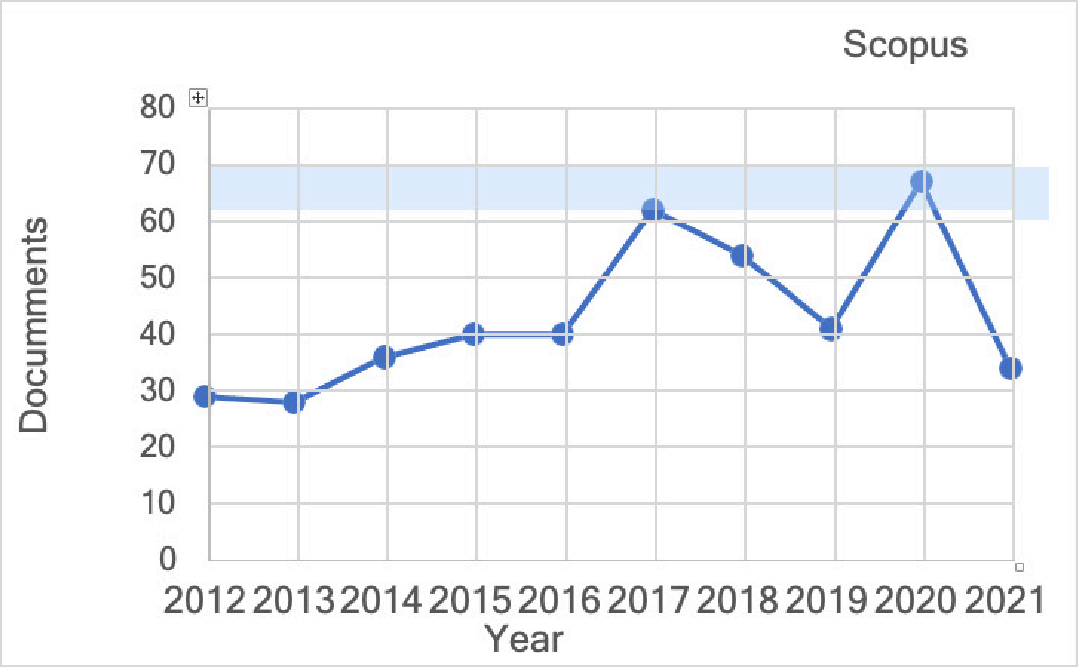
The increase in publications related to the words that were used for the two databases, have similar statistical trends towards an increasing polynomial mathematical regression, but it hasnot been well defined in the last five years. Although between 1984 and 2008 the production of articles on butanol was very low (Niemistö *et al*., 2013), since 2012, there has been a constant growth (Figure 1).

Perhaps due to the improvement of production costs and an increased interest in other competitive substances. Likewise, it seems that the number of papers per year is supported by the focus on research with new raw materials or availability and advances in biotechnology (Trindade and Santos, 2017; Da Silva Trindade and Dos Santos, 2018; Lu *et al*., 2021).

The bibliometric analysis shows that from the types of documents related to butanol, 74 % are conventional research articles, 8,4 % are reviews; 6,9 % are section documents, 6,0 % are book chapters; 1,4 % are notes, 1,2 % are editorials, and 1.2% are other types of analysis. In Scopus database, the document with the highest citation is a review entitled "¿Challenges in butanol production: How to improve the efficiency?" (García *et al*., 2011). For the first semester of 2021 there are 43,5 % of documents published on Scopus. The possible tendency to increase is also evident in the WoS platform.

Evaluating the classification of areas made by Scopus and Web of Science platforms (Table 1), the greatest contribution corresponds to chemical engineering and engineering in general. The field of engineering contributes with 39 %, biological sciences with 31 % and environmental sciences with 23 % approximately. Other areas with lower contribution have tangential relationships with research in butanol applications. For example, in immunology, recent researchuses this compound to obtain natural active polysaccharides with immunomodulatory activity (Wang *et al*., 2021).

The area of biotechnology and energy shows a good contribution result, focusing on obtaining butanol for industrial and energetic application. For example, the use of fluorescent reporter proteins of acetogens, for the recombinant production of biological products such as butanol andacetone.



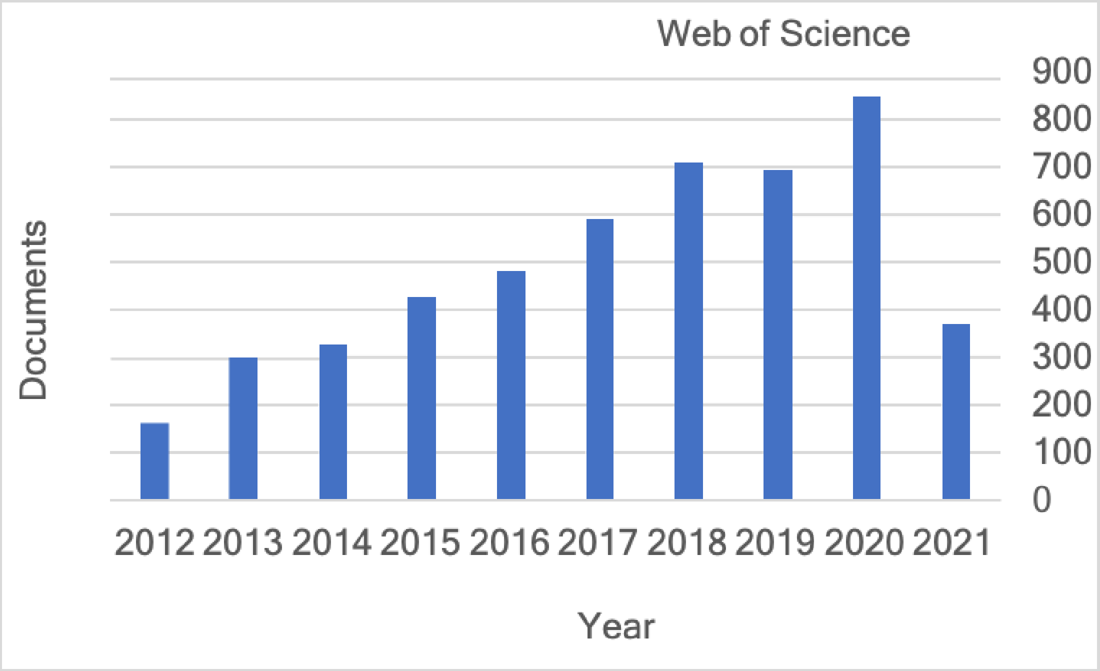


Figure 1. Evolution of published documents about butanol on databases.

Table 1. Documents by area of research

Scopus		Web of Science	
Thematic area	**%. Contribution**	**Thematic area**	**% Contribution**
Chemical Engineering	23,2	Agriculture	28,5
Energy	16,2	Biotechnology AppliedMicrobiology	18,5
Environmental Science	13,2	Energy Fuels	17,2
Chemistry	10,1	Engineering	15,8
Biochemistry, Genetics andBiology	9,3	Chemistry	12,3
Engineering	8,3	Environmental Sciences Ecology	11,9
Agricultural and BiologycalSciences	6,5	Science TechnologyOther Topics	9,6
Immulogy and Microbiology	4,6	Plant Sciences	8,2
Areas others	8,7		5,6

Source: www.scopus.com; www.webofscience.com

In addition, there is a reduction of greenhouse gas emissions (Flaiz *et al*., 2021), the biodegradation of the cell wall in sugar cane using small doses of an enzyme cocktail with *Fusarium metavorns* (Brandt *et al*., 2021), the detection of butanol on a photocatalytic silver (Ag) surface, used in the field of catalyst modeling and engineering (Devasia *et al*., 2021) and the controlled combustion of butanol and gasoline mixtures (Lu *et al*., 2021).

The statistical analysis of Top 10 document registers both in Scopus and Web of Science platforms (Table 2), as well as in VOSviewer, showed that the author with the highest number of documents published on the topic of butanol is Bonomi A. The countries that publish the most onthis topic are the United States and China, although Latin American countries like Brazil and Colombia are beginning to rank well.

Table 2. Main contributions on butanol research.

	Authors	Country	Sponsor	Author	Country	Sponsor
1	Bonomi, A.	United States	European Commission	Sun RC	Brazil	Conselho Nacional De Desenvolvimento Cientifico E Tecnologico Cnpq
2	Dias, M.O.S.	China	Fundação de Amparo à Pesquisa do Estado de São Paulo	Comstock JC	United States	Coordenacao De Aperfeicoament o De Apessonal De Nivel Superior Capes
3	Mariano, A.P.	Brazil	National Natural Science Foundation of China	Li, Y., Zhao, Y., Ishak, S. et al	Peoples R China	Fundacao De Amparo A Pesquisa Do Estado De Sao Paulo Fapesp
4	Lareo, C.	India	Conselho Nacional de Desenvolvimento Científico e Tecnológico	Li YR	India	National Natural Science Foundation Of China Nsfc
5	Marzocchella, A.P.	Canada	MinistÃ©rio da CiÃªncia, Tecnologia e InovaÃ§Ã£o	Kumar S	Spain	European Commission
6	Pereira, L.G.	South Korea	National Science Foundation	ViswanathaN R	France	United States Department Of Energy Doe
7	Yang, S.T.	Netherlands	European Commission	Que YX	England	National Science Foundation Nsf
8	Arakaki, N.	Spain	National Research Foundation of Korea	Singh S	South Africa	United States Department Of Agriculture (USDA)
9	Ferrari, M.D..	Colombia	Coordenação de Aperfeiçoamento de Pessoal de Nível Superior	Kumar R	Germany	Fundacao De Amparo A Pesquisa Do Estado De Minas Gerais Fapemig
10	Maciel Filho, R.	United Kingdom	U.S. Department of Energy	Zhang Y	Japan	Uk Research Innovation Ukri

Source: www.scopus.com; www.webofscience.com

According to the thematic network generated by VOSviewer, in butanol production from 1984 to the first period of 2021 (Figure 2, Table 3), a great diversity of topics has been worked on.

Table 3. Keyword cluster – butanol, 1984 to 2021

Cluster	Keywords (50 items)
Cluster I	22 items: alcohol production, article, bacterium, bagasse, biorreactor, bioreactors, butanol, butanols, chemistry, clostridium, clostridium Acetobutylicum, clostridium beijerinckii, controlled study, enzyme activity, glucosa, hydrolysis, metabolism, nohuman, priority journal, saccharum, sugarcane, unclassisfiend drug
Cluster II	15 items: alcohol, biodiesel, bioenergy, bioethanol, biofuel, biofuel production, biofuels, biomass, biorefineries, biorefinery, biotechnology, carbon, hydrogen, refining
Cluster III	10 items: acetic acid, acetone, butenes, computer software, distillation, economic analysis, etanol, fermentation, sugar, sugar cane
Cluster IV	3 items: cellulose, lignin, lignocellulose

Source: VOSviewer.exe software, versión 1.6.14.0

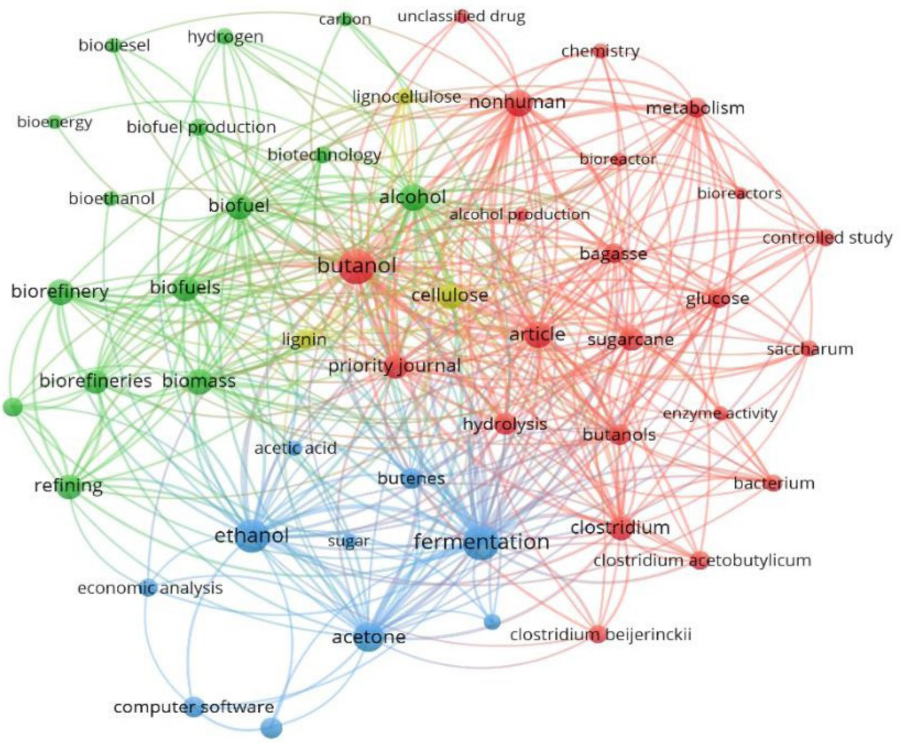


Figure 2. Network of topics related to butanol.

The topics of interest shown in the network by node size are: biofuel (Sarathy *et al*., 2018), glucose, *clostridium* (Groeger *et al*., 2017; Liberato *et al*., 2019), bacteria, ethanol (Nanda *et al*., 2017), acetone, hydrolysis (Xia *et al*., 2020), computational tools (Lozano-Parada *et al*., 2018; Bennamoun *et al*., 2020), biorefineries (Farzad *et al*., 2017; Mandegari *et al*., 2017; Wagemann& Tippkötter, 2019), economic analysis, biomass articles in general (de Mello *et al*., 2019) and sugarcane bagasse (Aitken *et al*., 2018; Vera-Gutiérrez *et al*., 2019; Pratto *et al*., 2020). The node of sugarcane bagasse appears small, distant from large nodes and without defined relationships

Available biomass for butanol is strongly related to cellulose, lignin and hemicellulose (Saini *et al*., 2015; Baksi, Saha, *et al*., 2019; Wu *et al*., 2018), and also with delignification and hydrolysis processes (Baksi, Sarkar, *et al*., 2019; Bhatia *et al*., 2020; Zheng *et al*., 2015). In the same way, microorganisms, genes and bacteria, are subjects that are worked through metabolic pathways, to improve biotechnological processes (Cimino *et al*., 2018).

In fermentation, the behavior of microorganisms depends on operating conditions such as: substrate concentration, nutrients, pH and temperature, among others (Goelzer & Fromion, 2011). The trend has been to look for more tolerant microorganisms and metabolite producers,in order to focus on those that are best marketed (Mariano *et al*., 2013).

Butanol is also related to fermentation, acetone and ethanol (Zheng *et al*., 2015; Pyne *et al*., 2016). These products are part of the ABE process (Lim *et a*l., 2020). The most recent records cover improvements in metabolic engineering (Xu *et al*., 2017; Feng *et al*., 2018), employing *Clostridium* bacterial species (Ibrahim *et al*., 2017) such as *beijerinckli*, but glycerol and 1,3- propanediol are little considered (Zheng *et al*., 2015; Pyne *et al*., 2016).

The major challenges for improving butanol production are oriented towards the development of genetic engineering, metabolic accuracies of microorganisms, conditions and integration of process operations, raw materials, and techniques for separation and purification of butanol afterfermentation (Kumar, M. and Gayen, K., 2011; ASOCAÑA, 2020-2021; Hoang *et al*., 2021; Sinumvayo *et al*., 2021). Product formation depends on all these parameters, which are permanently evaluated.

Clostridium genus microorganisms have shown significant activity in biobutanol production. Particularly the bacterium Clostridium pasteuriaum differs from other strains because of its capacity to fix nitrogen as well as to propagate in a carbon-rich medium, forming by-products. When in the presence of glycerol, it forms 1,3 propanediol (Pyne *et al*., 2016; Aragon, M. *et a*l., 2018), acetic acid, lactic acid, succinic acid, carbon dioxide and hydrogen (Xue *et al*., 2016; Groeger *et al*., 2017). In other words, in its fermentative action, it can generate metabolites with good added value (Dabrock *et al*., 1992; Kumar & Gayen, 2011; Zheng *et al*., 2015; Lipovsky *et al*., 2016). When fermentation is integrated with different extractions, there can be higher productivity and even butanol energy savings (Zhang *et al*., 2013).

With *C. beijerinckii* bacteria, final concentrations in the fermented solution between 21 and 27 g/L have been reported for barley and wheat straw, and corn stover (Kumar & Gayen, 2011). Although with corn stover, assuming a reactive efficiency of 90 % of fermentable sugars, it´s possible to have yields of 41 % butanol with respect to those sugars by simulation (Baral, N.R. *etal*., 2018). But with post-harvest sugarcane residues, in fermentations with *C. Saccharoperbutylacetonicum* DSM 14923, butanol concentrations of only 4,95 g/L and 0,65 to 5,93 g/L have been obtained with *C. Saccharobutylicum* DSM 13864. The latter data is more congruent with research conducted from hydrolysates with concentrations of 34 g/L of glucose and 22 g/L of xylose (from biomass with a lino-cellulosic purity of 79 %), where concentrations of 10,33 g/L of ABE (acetone, n-butanol and ethanol) were obtained, in the fermentate (Magalhães *et al*., 2018).

GRUBIOC obtained butanol concentrations in fermentates of 11,37 g/L with hydrolysates of sugarcane leaves and buds using *C. Pasteurianum*, (Aragón, M. *et al*., 2018). In continuous fermentations of sugarcane bagasse, butanol weight ratios per used substrate have been 0,22to 0,26 and in discontinuous fermentations from 0,28 to 0,33 (Michailos *et al*., 2016).

In biometrics, biorefineries with butanol show strong relationships with biofuels and biomass. However, in this case, this raw material is not very well linked, nor are the simulations, and even less so biogas, glycerol and microalgae; although lignocellulosic residues such as sugar cane have a strong relationship with them (Moral-Muñoz *et al*., 2020; Lu *et al*., 2021). In general, butanol research is focused on improving the performance and productivity of pretreatment operations, fermentation; compound separation and purification (Brandt *et al*., 2021).

The disadvantages of the ABE (Acetone, buthanol and ethanol) process are also considered in operational, energy and environmental costs. For example, the cost of substrates from petroleum, the high energy in butanol recovery, low yields (due to the metabolism and the typeof the microorganism), high water consumption and its treatment in WWTPs (Dürre, 2011).

In biorefinery simulation studies to produce ABE, it has been reported an overall butanol production yield of 52 % (Furtado-Júnior *et al*., 2020). Through biorefinery simulation it was possible to obtain from coffee waste, a yield of 140 kg of ABE per ton of coffee stalks (Carmona- Garcia *et al*., 2019). Studies begin with first generation feedstocks, mainly from sugarcane, to obtain ethanol, sugar and n-butanol, where they have focused, in addition to technical feasibility, economic, environmental and social sustainability, under the context of circular bioeconomy (Farzad S. *et al*., 2017; Mandegari *et al*., 2018).

Biofuels such as ethanol and biodiesel have been studied in depth, but butanol not so much (Martínez *et al*., 2020). The permanence of the ABE process is evident since its production was first initiated. The evolution has been slow due to high production costs and the emergence of petrochemicals. Although in the 1960s, the increase in the price of substrates caused its collapse and emergence (Durán-Padilla, 2015), but then in the 1990s, with the advances in science and technology, butanol production improved.

Through sustainability, economic and energetic analyses, using tools such as Aspen, GAMSand SimaPro, studies have been made from sugarcane bagasse to obtain methanol and butanol biochemically. Multi-criteria decision analysis methods (MCDA) have been applied and as wellas kinetic models in the integration of pyrolysis and gasification operations (Michailos *et al*., 2016).

From 2008 to 2020, biotechnological processes have again become relevant by a significant increase of the number of papers published. The topics that were addressed correspond to advances in metabolic engineering, genetics, biosynthesis, use of technical economic analysis, pretreatment and cellulosic butanol separation technologies. For the first half of 2021, research on proteins for the recombinant production of biological products, advanced bio-alcohol detection techniques and effective combustion control of improved butanol/gasoline blends are reported. These new insights are opportunities to scale up lignocellulosic butanol production witha biorefinery approach.

Profitability of n-butanol is analyzed in a review paper on butanol characteristics, production and use as a fuel in internal combustion engines (Trindade y Santos 2017. International prices as of 2016 for ethanol (0,66 $US/L), chemical n-butanol (1,34 $US/L), fuel n-butanol (0,83 $US/L), sugar (0,48 $US/kg) and sugarcane (27,26 $US/t) are taken into account. The study concludes that biobutanol is more profitable for the chemical industry as a commodity than as fuel, but it is lacking a biorefinery approach.

Regarding different scale biorefineries, optimizations have been considered regarding hypotheses of real economic precision calculations in which the plant operates (Belletante *et al*. 2020), with multiple feedstocks to produce ethanol, butanol, succinic acid, levulinic acid and hydrogen (Meramo-Hurtado *et al*., 2020). The evaluation analyses by simulation, are technical- economic (TEA) and/or life cycle (LCA) (Baral, N.R. *et al*., 2021; Meramo-Hurtado *et al*., 2020; Furtado Júnior *et al*., 2020; Michailos *et al*., 2016; Ling Tao *et al*., 2013; Ginni *et al*., 2021; Hao,J. *et al*., 2021).

CONCLUSIONS

Corn, rice, rye, wheat, sorghum, banana, cocoa, cassava and wood, have been the moststudied lignocellulosic residues for obtaining butanol, but the most complete research is relatedto sugar cane. The bibliometric review on butanol research, indicates that from the year 2000 onwards, publications started to increase exponentially, particularly looking for lignocellulosic biomass as feedstock. Although publications on the subject are not many, compared to other biofuels (mainly ethanol and biodiesel), the leadership of the United States and China; followed by Brazil and Colombia, is vast. Biotechnological processes still show certain weaknesses, particularly fermentation processes, due to the difficulty of finding microorganisms tolerant to the metabolites involved and to wider normal thermal conditions than environmental ones. It would be also necessary to know in greater depth, the metabolic pathways of these microorganisms and to involve the biorefinery approach much more, with more aggregate products that could be obtained. There are no detailed studies where the technological impacts integrated with environmental, economic and social impacts can be well visualized. Perhaps because of the lackof data, especially experimental data.

DECLARATION OF COMPETING INTERESTS

The authors declare that they have no known competing financial interests or personal relationships that might appear to influence the work presented in this bibliometric analysis.

ETHICAL APPROVAL

Not applicable

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AUTHORS’ CONTRIBUTIONS

DMM analyzed the data and wrote the manuscript and JELG participated in writing the manuscript and revised the paper. All authors read and approved the final manuscript.

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